Stopping Powers of Al, Ti, Fe, Cu, Mo, Ag, Sn, Ta, and Au for 7.2 MeV Protons (Memorial Issue Dedicated to the Late Professor Yoshiaki Uemura)

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Stopping Powers of Al, Ti, Fe, Cu, Mo, Ag, Sn, Ta, and Au for 7.2 MeV Protons

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and
the late Yoshiaki UEMURA**

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Stopping powers of Al, Ti, Fe, Cu, Mo, Ag, Sn, Ta, and Au for 7.2 MeV protons have been measured using a silicon detector. It has been confirmed that the present results for Al, Cu, Ag, and Au are in good agreement with the previous work. It has been found that Nara data are 1.5~3 percent lower than the data of Andersen *et al.* It appears that the deviations are decisive. It has been discussed that Nara data accord with the range data of the compilation of Whaling and of Rybakov as well as the stopping power data of Burkig and MacKenzie at 20 MeV in the absolute scale. It has been also shown that Nara data accord well, as a whole, with the tables of Barkas and Berger. Some remarks have been given on Andersen's experiment and Nara experiment. The Oscillatory behavior of Bloch constant with increasing Z has also been discussed.

I. INTRODUCTION

The accurate data of the stopping power of various materials for heavy charged particles are necessary in many fields of physics such as nuclear physics, radiology, biological physics and health physics and so on.

In 1964, when a compilation[1] of extensive review works on the penetration of charged particles in matter was published, Bichsel[2] made a critical review of experimental stopping power and range data for heavy charged particles. It turned out that the accuracy of informations given by the existing data at that time was not much better than 5 percent except for the case of Al.

More recently, extensive measurements of the stopping powers of metalic elements were performed by Andersen *et al.*[3~6] The calorimetric method was used to determine the energy loss of the particles in the sample foils. Their results have been presented as the stopping power tables for protons from 2.25 to 12 MeV. The accuracy of their results has been stated to be 0.3 percent.

On the other hand, the stopping powers of Al, Ni, Cu, Rh, Ag, Pt, and Au for 7.2 MeV protons and 14.4 MeV deuterons were measured using a surface barrier type silicon detector.[7] The stopping power data for protons were compared with the data of Andersen *et al.* (henceforth abbreviated to Andersen), and it was found that Nara data are some 1~2 percent lower than Andersen's data. Since the uncertainty of Nara data has been estimated to be about 0.5 percent, the deviations should be regarded as

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statistically significant.

Two possibilities of the systematic error which might give rise to the too low stopping power value in our experimental procedures have been investigated. One is the problem of the base line shift of the amplifier system and the other is the effect of the goodness of the geometry in our experimental setup. The former possibility has been denied in the addendum of the previous paper.[7] The latter has been investigated experimentally using Po^{212}(ThC') alpha particles and has also been rejected. This experiment will be published elsewhere.

Thus, the deviations between Nara data and Andersen's data have remained unexplained. In view of the overwhelming amount of Andersen's data and their great influence, it was considered indispensable to remeasure the stopping power for protons, even if it were at one fixed energy, with a method other than calorimetric one and of sufficiently high accuracy. In the present work, the stopping powers of Al, Ti, Fe, Cu, Mo, Ag, Sn, Ta, and Au for 7.2 MeV protons have been measured using a surface barrier type silicon detector with quite the same procedures as in the previous work.[7] However, the energy calibration procedures for the present work have been performed quite independently of the previous work.

II. EXPERIMENTAL PROCEDURE

1. Experimental Setup.

The experimental setup to determine the energy loss of protons in the sample absorber is quite the same as described in detail in the previous paper.[7]

The experimental setup is shown in Fig. 1. The molecular hydrogen ions accelerated to 14.4 MeV with the Kyoto University cyclotron were used. To convert the molecular hydrogen ions into protons, an aluminium foil of about 7 microns was inserted to the beam just before the object slit S_1 of the analyzing magnet.

The beam scattered at an angle of 15 degrees by a gold scatterer of 180 $\mu g/cm^2$ was used for the measurements. The sample foil was fitted to one of the windows of the absorber wheel and the wheel was rotated in front of a silicon detector (ORTEC). Thus, the pulse height with and without the absorber foil was measured simultaneously in one exposure. The pulses from the detector were amplified with a low noise amplifier. The relevant portion of the pulse height spectrum was expanded by a biased amplifier and fed into a 400 channel pulse height analyzer. From the pulse height difference with and without the absorber, the energy loss of protons in the sample foil was determined.

2. Determination of Incident Beam Energy.

Since the deviation of the absolute value of the stopping power (henceforth abbreviated to S value) is the question at issue, the procedures of energy determinations will be described somewhat in detail.

The energy of the protons was absolutely determined by the analyzing magnet. The momentum resolution of the analyzing magnet was set to be 0.1 percent. The magnetic field was stabilized by a current stabilizer and was measured by the method of nuclear magnetic resonance. During the time of exposure from 10 to 20 minutes,

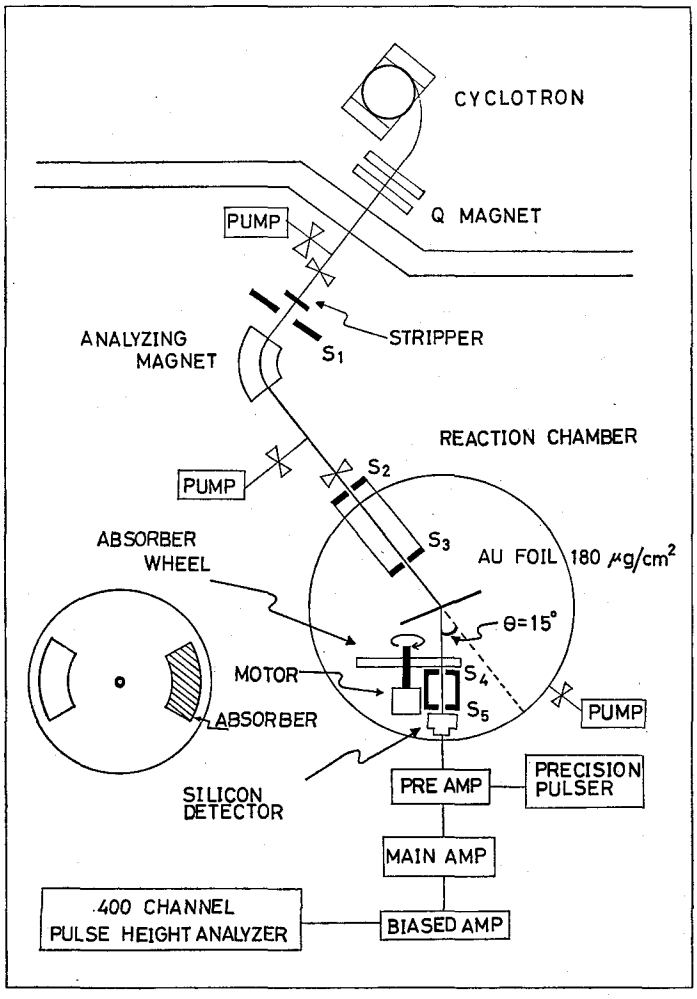


Fig. 1. Experimental setup for the energy loss measurement.

the magnetic field was kept constant within 0.01 percent. Accordingly, the central value of the analyzed beam energy was kept constant better than 0.02 percent throughout the experiments.

Since the radius of curveture of proton trajectories in the analyzing magnet depends on the lateral displacement of the slit S_1, the analyzing magnet was calibrated for the actual slit setting of the present experiment using Po^{212} alpha particles. In Fig. 2 the transmission curve for Po^{212} alpha particles is shown. In this measurement the width of the slit S_1 was doubled (the center was unchanged), because the intensity of the Po^{212} source was much weaker than accelerated protons. And the counts were corrected for the decay.

From Fig. 2 the average frequency was determined as

$$<f> = 23.0452 \pm 0.0012 \text{ MHz}.$$

Using Ritz's value[8] for the magnetic rigidity of Po^{212} alpha particles, the effective radius of curveture of the proton trajectories in the analyzing magnet was determined as

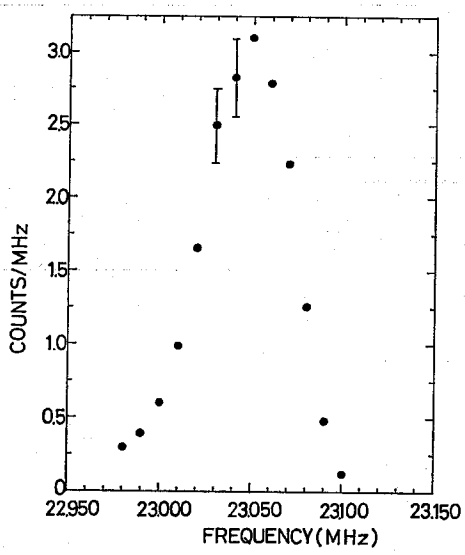


Fig. 2. Transmission curve for Po^{212} alpha particles of the analyzing magnet.

$$\rho = 78.899 \pm 0.006 \text{ cm.}$$

In the actual S value measurements the magnetic field was set at 20.9760 ± 0.0008 MHz. This corresponds to the proton energy of 7.2092 ± 0.0011 MeV. This is the energy of protons incident on the gold scatterer. The energy of protons scattered at the angle of 15 degrees by the gold scatterer and incident on the sample foil was calculated by using relativistic kinematics and exact mass values.[9] In order to obtain the maximum resolution, the gold scatterer was mounted in such a way that the normal to the scatterer was at an angle of 7.5 degrees (one half the scattering angle) with respect to the incident beam direction. It was assumed that the energy of the proton scattered at the median point of the scatterer approximates the central energy of protons incident on the sample foil. The energy absorption in the gold scatterer was calculated by the quadratic interpolation of Andersen's table.[4] The resultant energy is 7.2026 ± 0.0012 MeV.

3. Calibration of Pulse Height Spectrum.

Since only the relevant portion of the entire pulse height spectrum was expanded by the biased amplifier and recorded on 400 channel pulse height analyzer, it is necessary to calibrate the pulse height spectrum in energy. For this aim the pulse heights of protons elastically scattered by an aluminium foil of 1.692 mg/cm^2 at various angles were recorded. The energy of protons at each angle was calculated by quite the same procedure as in the case of gold scatterer. The energy absorption in the aluminium scatterer was estimated using Bichsel's table.[10] The energy scale was cross checked by a precision pulser. The pulser was normalized by the detector pulse without the absorber, *i.e.* protons of 7.2026 MeV.

The advantage of the method which utilizes elastic scattering of protons consists in the fact that the ionization defect in the energy responce of the detector and the effect

of the dead layer at the surface of the detector are automatically eliminated.

In the calculation of the present work, the physical constants of 1969[11] were used.

4. Sample Foils.

All foils were rolled ones. Square samples of 2 cm by 2 cm were cut out with a razor's blade. Each foil was weighed on Metler M–5 microbalance five times. The area of each foil was measured with Tiyoda LTG bi–A II microscope which can read to 1 micron five times.

The repetition of the whole set of weight per area measurements for the same sample at different date showed the standard deviation of 0.1 to 0.15 percent. Therefore, the uncertainty of the foil thickness was assigned to be 0.15 percent for all foils.

Thickness, stated purity and supplier of each foil are shown in Table I.

Table I. Thickness, Stated Purity and Supplier of the Foils. The Uncertainty of the Thickness is 0.15%.

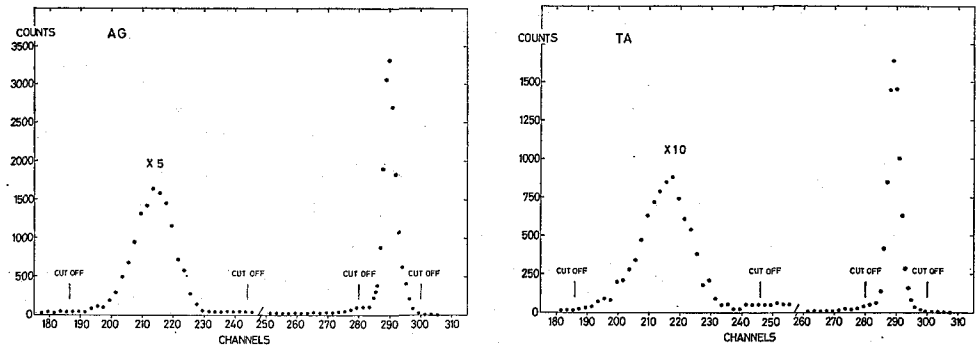
Element	Thickness (mg/cm^2)	Purity (%)	Supplier
Al*	10.118	99.8	Toyo Al.[a]
Ti	9.222	99.5 (0.25% Fe)	MacKay[b]
Fe	12.424	99.9	Fukuda[c]
Cu*	14.840	99.9	Fukuda
Mo	13.716	99.95	MacKay
Ag*	17.737	99.9	Fukuda
Sn	14.183	research grade	Unknown[d]
Ta	21.862	99.99	Fukuda
Au*	20.379	99.95	Ishifuku[e]

a) Toyo Aluminium Co., Ltd.
b) A. D. MacKay Inc.
c) Fukuda Metal Foil and Powder MFG Co., Ltd.
d) Supplied through Institute of Nuclear Study, University of Tokyo.
e) Ishifuku Metal Industry Co., Ltd.
* *The very same foil as used in the previous work.*

III. RESULTS

The measurements were made four times for each elements Al through Sn. For Ta and Au, the measurements were made five times. Typical pulse height spectra are shown in Fig. 3. The peaks with absorbers show nearly symmetrical distributions for all elements. The pulse heights were determined by taking the average value. To avoid the ambiguity caused by the choice of the cut-off point of averaging, the range of averaging was chosen sufficiently large. By changing the cut-off points by 5 channels step on both sides, the average value was confirmed to change equal to or less than 0.1 percent of the pulse height difference between the peaks with and without the absorber.

The numerical results of the pulse height measurements for Cu and Ta are shown in Table II as examples. The standard deviation (not standard error) of the pulse



(a) Ag (b) Ta

Fig. 3. Typical pulse height spectra.

Table II. Examples of Numerical Results of Pulse Height Measurements. P.E. Denotes the Propagation Error and S.E. Denotes the Standard Error.

Element and Run	No Absorber Peak (channel)	Absorber Peak (channel)	Pulse Height Difference (channel)
Cu Run 1	289.874±0.025	213.383±0.119	76.491±0.121
Run 2	290.354±0.024	213.723±0.099	76.631±0.102
Run 3	290.433±0.024	213.821±0.101	76.611±0.104
Run 4	290.394±0.022	213.949±0.102	76.444±0.104
Average	290.263±0.012 (P.E.) ±0.131 (S.E.)		76.544±0.054 (P.E.) ±0.044 (S.E.)
Ta Run 1	290.330±0.032	214.825±0.277	75.506±0.279
Run 2	290.083±0.028	215.235±0.223	74.849±0.225
Run 3	290.338±0.029	215.204±0.217	75.135±0.219
Run 4	290.425±0.027	214.892±0.203	75.532±0.205
Run 5	290.357±0.027	214.880±0.205	75.477±0.206
Average	290.307±0.013 (P.E.) ±0.058 (S.E.)		75.300±0.102 (P.E.) ±0.136 (S.E.)

heights of no absorber peak for 38 times measurements in all was 0.430 channels. As the integral pulse height corresponds to about 1067.4 channels, the stability of the amplifier system was 0.040 percent during about one week experiments.

As in the present experiments the pulse heights with and without the absorber were recorded simultaneously in one exposure, the pulse height difference is affected by the gain drift of the amplifier system only about 0.04 percent or less. This is by far the smaller than other uncertainties. No correction was made for the gain drift of the amplifier system.

The energy calibration measurements by scattering by the aluminium foil were made twice and the pulser measurement was made once. One of the scattering measurements is shown in Fig. 4. The 15° (Au) points were obtained from the no absorber peak for Au and Ta measurements respectively, which were performed just before each scattering measurement. By assuming the linear relation between the pulse height and the energy,

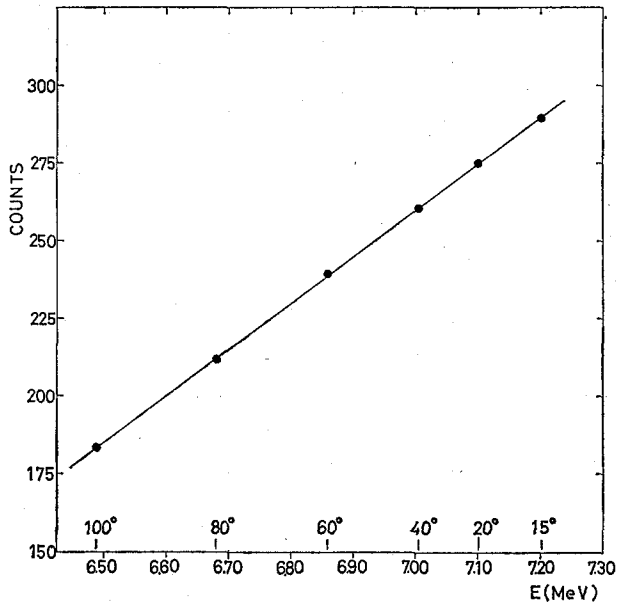


Fig. 4. The energy calibration by the scattering of protons.

the slope of the straight line, the energy value per channel, was calculated by the method of least squares. In pulser measurement, the ionization defect for 7.0 MeV protons was estimated to be about 10 keV from the measurement of Morton *et al.*[12] The weighted average of the two scattering measurements and one pulser measurement was 6.748 $\pm$0.025 keV/channel. The possible systematic error which might come out in the above determination of the slope was investigated and found to be at most 0.4 percent. But this is the extreme case and the possibility that such a systematic error really come out was considered to be very small. The attached error does not include the systematic error.

The energy loss in the sample foil was obtained by multiplying the above determined slope into the pulse height difference.

Since in the present experiment only protons which passed through the double slit system S_4 and S_5 (1.5 mm and 2 mm in diameter respectively and 80 mm apart) were detected by the silicon detector, the actual path length of protons in the foil was assumed to be equal to the foil thickness and no correction was made for the multiple scattering. The systematic error caused by this assumption is at most 0.05 percent.

The energy loss devided by the foil thickness, $\Delta E/\Delta t$, corresponds, in a good approximation, to the stopping power at the average energy defined by

$$\bar{E}=E_0-\Delta E/2\,,$$

where E_0 is the energy of protons incident on the sample foil.[3] The results are shown in Table III. It was assumed that the S value is proportional to $\ln v^2/v^2$ in a narrow velocity range. The present results have been reduced to 7.0 MeV by multiplying $(\ln v^2/v^2)_{7.0}/(\ln v^2/v^2)$. Since in the present experimental setup the proton beam does not scan all over the sample foil, an additional error of 0.2 percent has been added to

Table III. The Results. The Incident Energy Is 7.2026±0.0012 MeV. The Reduction to 7.0 MeV Was Made by Multiplying $(\ln v^2/v^2)_{7.0}/(\ln v^2/v^2)$.

Element	Δt (mg/cm^2)	ΔE (keV)	$\bar{E}$ (MeV)	$\Delta E/\Delta t$ (keV/mg cm^{-2})	$\Delta E/\Delta t$ reduced to 7.0MeV
Al	10.118	442.0	6.9827	43.68	43.58
	±0.015	±1.6	±0.0014	±0.19	±0.19
Ti	9.222	345.7	7.0309	37.49	37.65
	±0.014	±1.3	±0.0013	±0.17	±0.17
Fe	12.424	450.9	6.9783	36.29	36.18
	±0.019	±1.6	0.0014	±0.16	±0.16
Cu	14.840	516.5	6.9455	34.80	34.54
	±0.022	±1.8	±0.0015	±0.15	±0.15
Mo	13.716	410.8	6.9984	29.95	29.94
	±0.021	±1.6	0.0014	±0.14	±0.14
Ag	17.737	518.3	6.9446	29.22	29.00
	±0.027	±1.8	±0.0015	±0.13	±0.13
Sn	14.183	389.1	7.0092	27.43	27.47
	±0.021	±1.4	±0.0013	±0.12	±0.12
Ta	21.862	508.1	6.9497	23.24	23.08
	0.033	±2.0	±0.0015	±0.11	±0.11
Au	20.379	452.9	6.9773	22.22	22.15
	±0.031	±1.7	±0.0014	±0.10	±0.10

each uncertainty of the S value given in Table III. This additional error stands for the possible nonuniformity of the sample foils.

IV. DISCUSSION

1. Comparison of Present Results with Previous Results.

In Table IV the comparison of the present results with the previous results are shown for the common elements. All values have been reduced to 7.0 MeV. For Al, Cu, and Ag the agreement is very good. For Au the difference is larger than 1 percent

Table IV. Comparison with the Previous Results.* Both Present and Previous Results Have Been Reduced to 7.0 MeV.

Element	Present Results (keV/mg cm^{-2})	Previous Results (keV/mg cm^{-2})	Difference (%)	Weighted Average (keV/mg cm^{-2})
Al	43.58±0.19	43.67±0.25	−0.21±0.71	43.62±0.15
Cu	34.54±0.15	34.45±0.18	+0.26±0.67	34.50±0.12
Ag	29.00±0.13	28.96±0.18	+0.14±0.76	28.98±0.11
Au	22.15±0.10	22.46±0.18	−1.40±0.95	22.26±0.09

* Ref. 7).

but not significant statistically. Thus, the repeatability of our experimental procedures has been confirmed to be satisfactory. In the fifth column the weighted averages are given.

2. Comparison of Nara Data with Andersen's Data.

In Table V Nara data are compared with Andersen's data. Nara data for Ni and Pt are taken from the previous work. Again Nara data have turned out to be lower than Andersen's data by 1.5~3 percent. The deviations for all elements are statistically significant. It appears that the deviations for light elements are somewhat larger than for heavy elements. This trend was also seen in the previous work.

As already mentioned, two possibilities of the systematic error which might give rise to the too low S value in our experimental conditions were investigated and have been rejected.

Thus, the discrepancies between Nara data and Andersen's data appear to be decisive.

Table V. Comparison with Andersen's Data. All Data Have Been Reduced to 7.0 MeV. Data of Ni and Pt Are Taken from the Previous Paper.* Data for Al, Cu, Ag, and Au Are Average Values of Present and Previous Data.

Element	Nara Data (keV/mg cm^{-2})	Andersen et al.** (keV/mg cm^{-2})	Difference (%)
Al	43.62±0.15	44.81±0.13	−2.73±0.46
Ti	37.65±0.17	38.92±0.12	−3.37±0.55
Fe	36.18±0.16	37.28±0.11	−3.04±0.53
Ni	36.16±0.19	37.29±0.11	−3.13±0.61
Cu	34.50±0.12	35.12±0.11	−1.80±0.46
Ag	28.98±0.11	29.48±0.09	−1.73±0.48
Ta	23.08±0.11	23.66±0.07	−2.51±0.56
Pt	22.20±0.12	22.54±0.07	−1.53±0.63
Au	22.26±0.09	22.67±0.07	−1.84±0.51

* Ref. 7).
** Ref. 4), 5), 6).

3. Comparison of Nara Data with Data of Burkig and MacKenzie.

In order to examine the discrepancies between Nara data and Andersen's data, it will be wiser to compare these data with other experiments rather than to investigate the possibility of the systematic errors in both experiments.

We shall restrict our discussion within the energy range of the present interest. Sachs and Richardson[13~16] have performed absolute measurements of S values for various elements at proton energy of 18 MeV and derived I values, the mean excitation potentials. However, their experimental procedures are out-of-data and their I value for silver is conspicuously too high as compared with the I values of other elements. We shall except their data from the present discussion to avoid confusion, although their I values for Al, Cu, and Au agree well with those of later experiments.

The most reliable S value measurement is the experiment of Burkig and MacKenzie[17] (henceforth abbreviated to B–M). They have measured S value of 23

elements relative to Al for 19.8 MeV protons and derived I values for Be, Fe, Cu, Ag, W, Au, and Pb. In the same year, Bichsel, Mozley and Aron[18] have measured the ranges of protons from 6 to 18 MeV in Be, Al, Cu, Ag, and Au and derived I values. The I values derived in these experiments agree with each other. The Bloch constant, I/Z, thus obtained was 12.5~13 eV. On the basis of these experiments, Sternheimer[19] has calculated extensive tables of S values and ranges for protons up to 100,000 MeV.

After that, it has become recognized that the true Bloch constant is about 10 eV rather than 13 eV from the experiments[20,21] made at high energies, at which the shell correction should take the minimum value. It has been suggested[22,23] that the discrepancy between the Bloch constants obtained at low and high energies can be removed by the realization that the higher shell corrections are more important than they were considered to be.

In Table VI, are shown the I values derived by B–M[17] and Bichsel *et al.*[18] together with the presently accepted I values. It has been stated that the I values of B–M were obtained taking Bichsel's I value[18] for Al of 166.4 eV as standard and using Walske's shell corrections.[24,25] Accordingly, if the S values are calculated using the Bethe formula with I values of B–M and Walske's shell corrections, one can obtain the absolute values of S values for Al, Fe, Cu, and Ag*. We shall call them the "experimental S

Table VI. The I values of Burkig and MacKenzie, Bichsel, Mozley and Arom and Presently Accepted I Values (in eV).

	Al	Fe	Cu	Ag	Au
Burkig and MacKenzie	166.4	328.8	366.0	587.0	997
Bichsel *et al.*	166.45 ±1		375.6 ±20	585 ±40	1037 ±100
Presently Accepted I Values	163[a),b),d),e),f)] 166[e)]	273[d)] 283[e)]	312[e)] 315[d)] 320[b),c)] 322[f)] 326[b)]	471[d)] 475[e)] 480[e)] 485[f)]	761[d)] 775[e)]

a) Ref. 30).
b) Ref. 2), p. 17.
c) Ref. 26).
d) Ref. 1), p. 287., Ref. 22), p. 25.
e) Ref. 1), p. 99., Ref. 23).
f) Ref. 10).

value of B–M". If the internal consistency is retained, the relative magnitude of these calculated S values must agree with the originally observed relative S values. Inscrutably, however, the internal consitency does not hold for the I values of B–M.

In Table VII, are shown the experimental S values of B–M (calculated in the present work) for 20 MeV protons together with the S values obtained from various

* For Au, B–M took into account the higher shell corrections, the amount of which are not known.

Table VII. Burkig and MacKenzie's Experimental Stopping Powers and Stopping Powers from Various Tables at 20 MeV. Stopping Powers Are Given in keV/mg cm^{-2}.

	Al	Fe	Cu	Ag
Burkig and MacKenzie ("Experimental Values")	19.685	16.515	15.811	13.548
Burkig and MacKenzie (Original Relative Values)	1.000	0.856 ±0.002	0.821 ±0.002	0.715 ±0.003
Sternheimer	19.70		15.91	
Bichsel (2nd)[a]	19.765		16.235	14.039
Barkas and Berger	19.737	17.015	16.321	13.957
Janni	19.765	17.139	16.236	14.095
Serre	19.73	17.19	16.32	
Bichsel (3rd)[b]	19.690		16.237	14.134

a) Ref. 10).
b) Ref. 26).

tables.[10,26~29] As is obviously seen, the experimental S values of B–M do not agree with the originally observed relative S values. The origin of this inconsistency is incomprehensible, because no detail of the calculation of the I values has been described by the authors.

In the tables of Bichsel,[10,26] Barkas and Berger,[27] Janni,[28] and Serre,[29] the relative S value of Al, Fe, Cu, and Ag appears to agree fairly well with the observed relative S values of B–M.

It should be noted, first, that S values for Al in all calculations agree very well with one another. The difference is at most 0.4 percent.

It should be noted, secondly, that the relative magnitudes of the experimental S values of B–M for Fe, Cu, and Ag agree fairly well with the original relative S values. It appears that there exists a gap between Al and other three elements in the I values of B–M. At the same time, the experimental S values of B–M for Fe, Cu, and Ag are significantly lower than the values of various tables.

As the S value and the I value of Al up to 20 MeV have been considered to be well established,[30] tables of Bichsel, Barkas and Berger, Janni, and Serre appear to take the S value for Al as standard. Consequently, the S values for Fe, Cu, and Ag of these tables are of necessity higher than the experimental S value of B–M.

However, the experiment of B–M is the one and only measurement of S values at 20 MeV. And their I values for Cu, Ag, and Au agree with those obtained by Bichsel *et al.*[18] from the range measurements. Taking these reasons into account, it may not necessarily be the best choice to take the S value of Al as standard. On the contrary, it might be a better choice to take the experimental S values of Fe and Cu as standards. Although the origin of the gap between Al and other three elements in the I values of B–M is not clear, we shall try to take a standpoint in which the experimental S values of B–M for Fe and Cu are taken as standards.

In order to meet the original relative S values and retain the internal consistency,

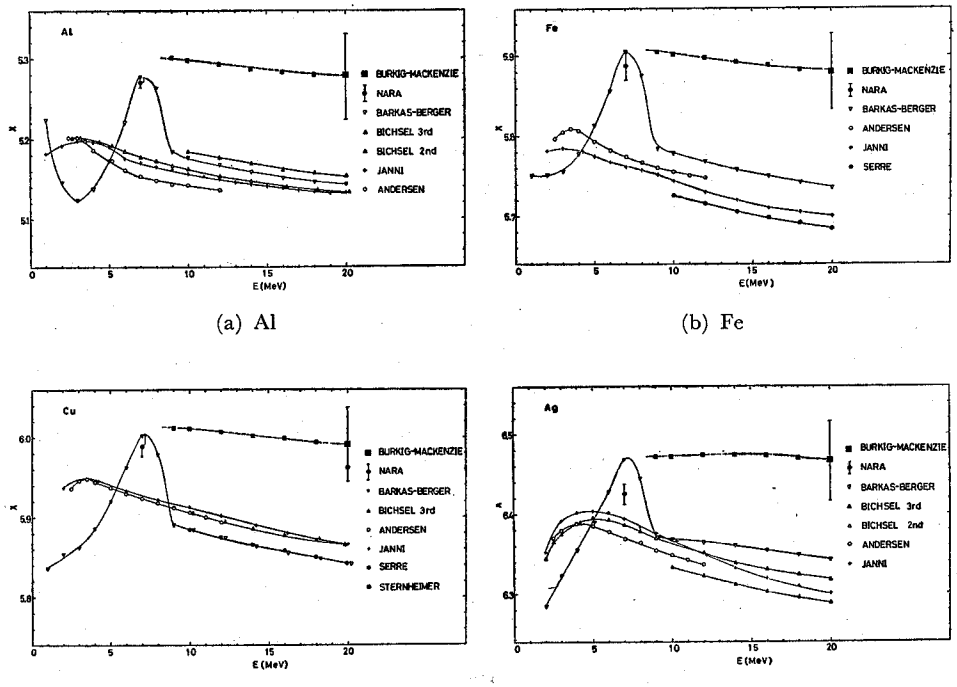
the S value for Al should be adjusted to be 19.25 keV/mg cm^{-2} instead of 19.685 (henceforth the unit of S value, keV/mg cm^{-2}, will be omitted)*.

In order to examine whether Nara data accord with the data of B–M or not, it is instructive to extract Bichsel's X value[2] from the S value and plot it against the energy. The X value is defined as

$$X = \ln I + C/Z$$

where I is the mean excitation potential in eV, C is the total shell correction and Z is the atomic number of the stopping material.

In Fig. 5, X values extracted from Nara data and from the experimental S values of B–M together with the X values from the various tables are shown. As stated by Barkas and Berger (henceforth abbreviated to B–B), their tables below 8 MeV are entirely empirical and the tables are based on the range data of protons for H_2, Be, and Al compiled by Whaling[31] and the range data for Fe, Cu, Sn, and Pb measured by Rybakov.[32] Above 8 MeV tables of B–B are based on the Bethe theory with appropriate assumptions on I values and shell corrections. The sudden decrease of the curve of B–B between



(a) Al (b) Fe

(c) Cu (d) Ag

Fig. 5. X value plots. The error bars attached to the points of Burkig and MacKenzie correspond to the uncertainties in S values of ± 1 percent for Al, Fe, and Cu and ± 1.2 percent for Ag.

* From the relative S values it is expected: for Fe 16.515/0.856=19.293, for Cu 15.811/0.821=19.258, and for Ag 13.548/0.715=18.948. Placing the weight of 0.2 on the value 18.948, the weighted average was adopted as 19.25.

8 and 9 MeV is without doubt due to the factitious joining of the two calculations below and above 8 MeV which are essentially of different nature. Below 7 MeV X value curve of B–B reflects the systematic trend which stems from Whaling's and from Rybakov's experimental range data or otherwise the systematic trend which is proper to the range formula of B–B. Above 9 MeV the X value curve shows the energy dependence of the assumed shell corrections in the Bethe theory.

As already seen in Table VII, the experimental S values of B–M are some 2.5～4 percent lower than the values of various tables. Accordingly, the X values of B–M are higher than those of the tables. In order to estimate the behavior of the X values at lower energies, we took the following procedures. First we obtained the ratio of the experimental S value of B–M to the value of B–B at 20 MeV. Next, we multiplied this ratio into the S values at lower energies obtained from the table of B–B. Then X values were extracted from these S values. The X value curves thus obtained are shown by dashed curves in Fig. 5.

Of course, there is no theoretical reason for multiplying a constant factor into the S value at each energy. However, since I value is a constant for each element, X value curve represents the energy dependence of the total shell correction. As clearly seen from the figures, the dashed curves are never unnatural in shape as compared with other curves obtained from various tables.

For Fe, Cu, and Ag, the extention of the dashed curve join smoothly with the X value of B–B at 7.0 MeV, which is based on Rybakov's experimental range data.[32] In case of Fe and Cu, Nara data agree well with the values of B–B. This fact indicates that Nara data also accord well with Rybakov's range data as well as the experimental S values of B–M in the absolute scale.

In case of Al, the S value for B–M was taken as 19.25. The extention of the dashed curve also join smoothly with the X value of B–B at 7.0 MeV, which is this time based on the compilation of Whaling.[31] Nara value for Al also agrees with the value of B–B, therefore also agrees with the compilation of Whaling.

For the table of B–B above 8 MeV, it will be necessary to modify the assumptions on I values and shell corrections to join the X value curve smoothly with the empirical X values at 7.0 MeV. The modified table will give X value curves more or less similar to the dashed curves in Fig. 5.

In conclusion, in evaluating the experimental S values of B–M, the S values for Fe and Cu were taken as standards (*i.e.* I values given by B–M and Walske's shell corrections) and the S value for Al was adjusted to be 19.25 to retain the internal consistency. Then, it has been found that the experimental S values of B–M accord very well, as a whole, with Whaling's and Rybakov's range data at 7.0 MeV and also with Nara data at 7.0 MeV.

4. Comparison of Nara Data with the Tables of Barkas and Berger.

In Table VIII, entire Nara data* are compared with the tables of B–B. B–B state that their tables have significance of three figures, but we have assigned the

* In the previous work,[7] the thickness of the Rh sample was too thin and S value for Rh was less accurate than other elements and unreasonablly too low. We should like to withdraw Rh datum in Ref. 7.

Table VIII. Comparison of Nara Data with Barkas and Berger's Table. All Data Have Been Reduced to 7.0 MeV.

Element	Nara Data (keV/mg cm^{-2})	I_{adj} (eV)	Barkas-Berger (keV/mg cm^{-2})	Difference (Nara–B–B)/Nara (%)
Al	43.62±0.15*	163	43.56±0.22	+0.14±0.61
Ti	37.65±0.17	247.5	37.01±0.19	+1.70±0.66
Fe	36.18±0.16	285	36.01±0.18	+0.47±0.66
Ni	36.16±0.19**	304	36.20±0.18	−0.11±0.72
Cu	34.50±0.12*	314	34.30±0.17	+0.58±0.61
Mo	29.94±0.14	439	29.69±0.15	+0.84±0.70
Ag	28.98±0.11*	487	28.58±0.14	+1.38±0.62
Sn	27.47±0.12	516	27.13±0.14	+1.24±0.66
Ta	23.08±0.11	739	23.05±0.12	+0.13±0.69
Pt	22.20±0.12**	787	22.34±0.11	−0.63±0.72
Au	22.26±0.09*	797	22.32±0.11	−0.27±0.64

* Average values of present and previous data.
** Previous data; Ref. 7).

standard error of 0.5 percent. The overall agreement is satisfactory. Significant difference is seen for Ti and barely significant differences are seen for Ag and Sn. These differences will be discussed in the later section.

Since the tables of B–B are based on the experimental range data of Whaling and of Rybakov, Table VIII shows that Nara data are, as a whole, in good agreement with the range data of Whaling and of Rybakov.

5. Comment on Bichsel's and Janni's Tables.

Bichsel[10,26,33] has performed multiparameter least square calculations to obtain the most probable values of the key parameters in the Bethe theory, *i.e.* the mean excitation potential and the shell corrections, which accord best with the whole body of the existing S value and range data. Bichsel has presented extensive tables of S value and range for protons up to 1,000 MeV which we have already noted in Table VII.[10,26] Janni[28] has also calculated extensive tables in such a way that the tables accord best with all existing data.

As already seen in Table VII, the S values for Fe, Cu, and Ag at 20 MeV given by Bichsel's and Janni's tables are some 2.5~4 percent higher than the experimental S values of B–M. In these tables, the substance for Al is essentially based on the range measurements of Bichsel and Uehling.[30]

Exclusive of Andersen's experiments,[3~6] the direct S value measurements are rather scarce. Nakano *et al.*[34] have measured the relative S values for protons at 28.7 MeV, but in their paper no I values are derived and the general feature of their results appears to accord with that of B–M. The experiment of B–M is the one and only experiment for about 20 MeV protons except the experiment of Sachs and Richardson.[13~16]

Therefore, the large deviations between the experimental S values of B–M and the S values of Bichsel's and Janni's tables do not necessarily mean that values of B–M are too low. On the contrary, there is much possibility that the values given by the

tables are rather too high, because I values for Cu, Ag, and Au of B–M agree well those obtained by Bichsel *et al.*[18] from range measurements.

Since Andersen's data agree fairly well with Bichsel's and Janni's tables in general, it should be emphasized that the tables of Bichsel and of Janni are not necessarily on the line of Fano's program in the strict meaning, *i.e.* the difference between the Bloch constant of ~ 13 eV at low energies and the Bloch constant of ~ 10 eV at high energies can be removed by the understanding of the importance of the higher shell corrections.

For example, for 20 MeV protons Walske's shell corrections[24, 25] $(C_K+C_L)/Z$ for Cu is about 0.0888. If the difference between I value of B–M and the presently accepted values obtained from the experiments at high energies is actually due to higher shell corrections (Table VI), the amount of the higher shell corrections should be

$$\ln 366 - \ln 320 = 0.1343.$$

Then, the total shell correction should be

$$C/Z = 0.0888 + 0.1343 = 0.2231.$$

While the total shell corrections extracted from Bichsel's and Janni's tables are only 0.09337 for Ref. 10, 0.09766 for Ref. 26, and 0.09795 for Ref. 28. The same feature is also seen for Fe and Ag.

The values of total shell corrections adopted by Bichsel and by Janni are by far the smaller than the value expected from the difference of I values. Accordingly, Bichsel's and Janni's S values for Fe, Cu, and Ag are of necessity higher than the experimental S values of B–M by about $2.5 \sim 4$ percent.

This problem can also be considered from another point of view. If we rely on the experimental S value of B–M for Al (19.685) instead of Fe and Cu and then calculate the S values for Fe, Cu, and Ag by multiplying the originally observed relative S values, we can calculate the I values which are required to obtain these S values using Walske's shell corrections. Table IX shows the expected S values and required I values to obtain these S values.

Table IX. The Expected S Values and Required I Values. See Text.

	Fe	Cu	Ag
Expected S (keV/mg cm^{-2})	16.850	16.161	14.075
Required I (eV)	298.5	330.2	498.9

The required I values are much smaller than the values derived by B–M and are rather nearer to the currently accepted I values. This fact again indicates that there exists a gap between Al and other elements in the I values of B–M. However, it should be remembered that the I values of B–M for Cu, Ag, and Au agree well with the I values derived by Bichsel *et al.*[18] from the range measurements, although the L shell correction used were of different origin. Further, I values of Sachs and Richardson for Al, Cu, and Au derived by Caldwell[16] using Walske's shell correction also accord with the I values of B–M.

Returning to the standpoint in which the S values of B–M for Fe and Cu are taken as standards, the I value which is required to obtain S value of 19.25 for Al at 20 MeV is 187.21 eV if we use Walske's shell correction. If we use modified L shell correction suggested by Bichsel and Uehling,[30] $C_L = 1.5/E$, the required I value is 187.68 eV. These values are much higher than currently accepted values, especially the values[35, 36, 37] obtained at high energies. Since Al has only three electrons in the M shell, it may be difficult to account for this deviation by the contribution of M shell correction only. Perhaps L shell correction might be much larger than that estimated by Bichsel and Uehling. That is, if we take I value of 166 eV, the required total shell correction to obtain the S value of 19.25 is

$$C/Z = 0.1653,$$

while Walske's shell correction $(C_K + C_L)/Z$ is 0.0451.

6. Other Evidences in Literature.

There are other evidences which show that Andersen's data are higher than other experiments.

In the critical review of experimental stopping power and range data, Bichsel[2] has shown the smoothed Nielsen's[38] S values from 1 to 5 MeV. These Nielsen's data are lower than those of Andersen by 0.6~0.7 percent for Al and 0.8~1 percent for Ni. While the smoothed Nielsen's data agree very well with the range measurements in the same energy range obtained at Rice Institute.[39]

In the appendix of the second printing of the same review, Bichsel has compared the range measurements made at University of Southern California[40] with Andersen's data. In this comparison, Andersen's S values were integrated and reduced to range data. The reduced range data of Andersen are smaller than those of U.S.C. by 0.73 percent for Be, 0.98 percent for Al and 1.06 percent for Ag respectively. This means that Andersen's S values are too high as compared with the range data of U.S.C.

7. Remark on Andersen's Experimental Procedures.

Since Andersen's data are very extensive and have great influence, the ground of the experimental procedures should be steady and reliable.

It should be emphasized that in Andersen's method the energy dissipated as heat was actually measured instead of the energy loss itself. Many corrections such as thermal expansion correction, Coulomb scattering corrections, X-ray correction and δ ray correction should be applied to the raw data. Andersen states that in the worst cases the thermal expantion correction exceeded 1 percent, Coulomb scattering corrections were 0.5 percent, X-ray correction was 0.4 percent and δ ray correction was 0.5 percent. These corrections should be applied by just correct amount for ecah element at each energy. As the physical processes of these corrections are very involved, it is considered to be very difficult problems to find the "unique solution" for each of these corrections.

Furthermore, Andersen's experiments were made at liquid helium temperature. It is considered that some effects such as the difference of the lattice vibration or the energy distribution of conduction electrons between liquid helium temperature and room temperature might affect the process of the energy loss of protons. Close ex-

aminations should be made on these effects.

8. Remark on Nara Experiment.

As already mentioned, the possible systematic error which might come out in the determination of the slope is at most 0.4 percent. Such an error, even if it really exists, can not explain the deviations between Nara data and Andersen's data. The only remaining possibility of the systematic error in our experimental procedures may be the error in the determination of the incident energy. If the incident energy of protons were in reality higher than the value which we believe to be, the deviations might be explained. The deviation of the incident energy required to explain the deviations of S values should be about 200 keV. This is about 2.86 percent of the incident energy. To give rise to this amount of error in the energy, the momentum measurement in the analyzing magnet should be in error by about 1.43 percent. That is the error of the average resonance frequency in the calibration measurement should be

$$23.0452 \times 0.0143 = 0.3295 \text{ MHz}.$$

Such a large deviation is far beyond our imagination, because the entire transmission range was restricted to within 0.120 MHz as seen from Fig. 2.

Furthermore, it should be remarked that in Nara experiment the energy loss itself has been measured instead of the heat dissipation. In this sense, Nara data are much less ambiguous than Andersen's data.

9. Oscillatory Behavior of Bloch Constant.

In 1964, when a compilation of extensive review works on the penetration of charged particles in matter[1] was published, it was suggested[2,3] that the mean excitation potential, I, decreases smoothly with increasing atomic number, Z, of absorbing material.

Andersen[6] has reported that I values show an oscillatory behavior with increasing Z. More specifically, it was discussed that the Bloch constant, $I/Z = I_0$, as expresssed in the form

$$X_{\text{exp}} - \ln Z = \ln I_0 + C/Z$$

increases with increasing Z in the fourth period of the periodic table from the S value measurements for $Z = 20$ to 30.

Furthermore, from the measurements of S values of Zr, Ag, Gd, Ta, Pt, and Au, Andersen has suggested that the same trend can be seen also in the fifth and sixth periods.

In Figs. 6, 7, and 8, the comparisons of the value of $(X_{\text{exp}} - \ln Z)$ extracted from Nara data with those from Andersen's data at 7.0 MeV are shown.

Figure 6 shows the fourth period. Since Nara data are confined to Ti, Fe, Ni, and Cu, only Ti, Fe, Co, Ni, Cu, and Zn are picked up from Andersen's data. Apart from the difference of the absolute value, the general tendencies agree well with each other. These tendencies appear to agree with the theoretical calculations of I/Z by Chu and Powers[41] based on the statistical approach[42,43] to the energy loss problem. This tendency can also be expected from Table VIII in which S value for Ti is significantly higher than the value of B–B. And B–B have assumed that the Bloch constant decreases monotonously with increasing Z.

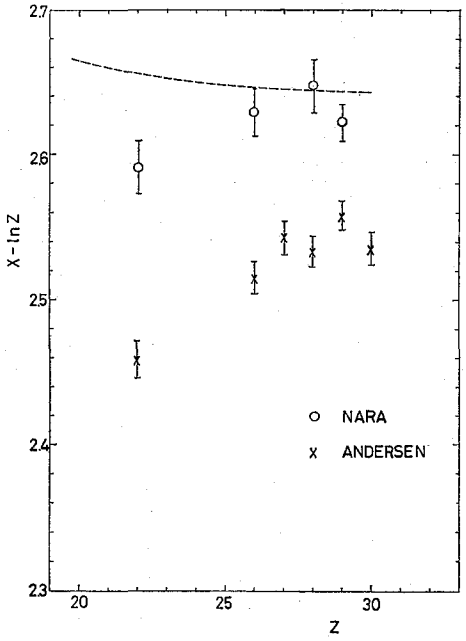


Fig. 6. (X_{exp}—ln Z) plot. Fourth period. The dashed curve represents the values extracted from the table of B–B.

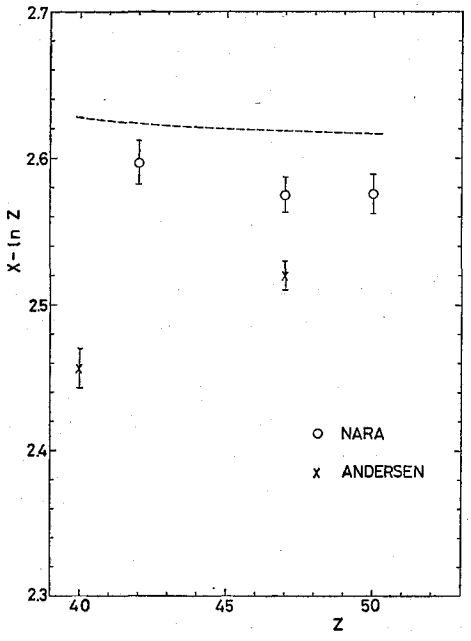


Fig. 7. (X_{exp}—ln Z) plot. Fifth period. The dashed curve represents the values extracted from the table of B–B.

Figure 7 shows the fifth period. Nara data comprise Mo, Ag, and Sn, while Andersen's data comprise Zr and Ag. In this case, the general tendencies do not agree with each other. In Nara data, no increase of $(X_{exp}-\ln Z)$ value with increasing Z are seen. The tendency of Nara data appears to agree with the experimental data of B–M and also with the theoretical calculation of Chu and Powers.[41]

Figure 8 shows the sixth period. Since Nara data contain only Ta, Pt, and Au, Andersen's data of Gd is omitted. Since available Nara data are confined to relatively short interval of the atomic number, the behavior of $(X_{exp}-\ln Z)$ value can not be judged. However, as for the three elements shown in the figure the general tendencies agree well with each other. In view of the scarcity of the observed points, the behavior of the $(X_{exp}-\ln Z)$ values in the sixth period can not be read from the presently available data.

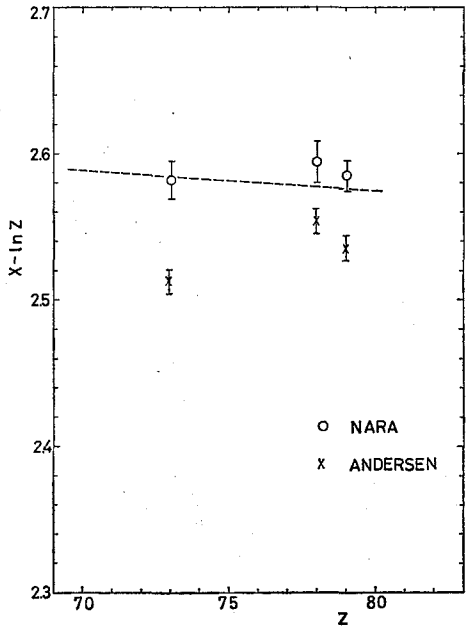


Fig. 8. $(X_{exp}$—ln $Z)$ plot. Sixth period. The dashed curve represents the values extracted from the table of B–B.

V. CONCLUSION

From the discussions made above it turned out that Nara data are decisively lower than the data of Andersen *et al.* However, it turned out that Nara data accord with the range data of Whaling and of Rybakov as well as with the stopping power data of Burkig and MacKenzie at 20 MeV in the absolute scale.

Since the stopping power data are most readily and unambiguously determined from the absolute energy loss measurement in relatively thin absorbers, it is most desirable to have the experimental energy loss data in the overlapping energy range by many different investigators and by methods other than calorimetric one.

VI. ACKNOWLEDGMENT

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Note Added in Proof.

Recently, Sørensen and Andersen (*Phys. Rev.*, **B8**, 1854 (1973)) have extended their measurements up to 18 MeV. With this extention stopping powers of Al, Cu, Ag, and Au at 7.0 MeV have been slightly altered. However, these alterations bring about no substantial changes in Table V.

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